The Empirical and Semi-Theoretical Methods For Predicting Viscosity of Binary n-Alkane Mixtures

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Abstract

In this study, the empirical and semi-theoretical new methods for viscosity of binary mixtures of n-alkanes are presented at atmospheric pressure and 288-333 K temperature ranges. In the new empirical viscosity calculation method, modified version of the Andrade equation and the simple mixture rule are used. The proposed new semi-theoretical method employs both the Enskog's hard sphere theory for dense fluids and the principle of corresponding states. The viscosities of binary mixtures of n-heptane with n-hexane and n-nonane covering different compositions were calculated using this method which requires only critical properties and normal boiling point as input data. The predictions were compared with excellent experimental data in literature. Highly satisfactory results were obtained. The percentage of average absolute deviations could be decreased to 1.2 and 0.9 utilizing the empirical and semi-theoretical viscosity methods, respectively, for 27 data points.

KEY WORDS: binary n-alkane mixtures, empirical viscosity calculation method, hydrocarbon, semi-theoretical viscosity prediction method, viscosity

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1. INTRODUCTION

Viscosity of pure organic liquids and their mixtures' data are indispensable for process design in engineering works, petrochemical and chemical industries. Unfortunately, reliable data on this important physical property are not abundant due to several experimental difficulties in its measurement [1-9]. However, several theoretical, semi-theoretical and empirical models for predicting the viscosity of pure components and mixtures are available in literature as excellent reviews[10-16].

All of the predictive and correlative viscosity methods of hydrocarbon mixtures use mixing rules. The mixture rules were used extensively by well known Mc Allister, Tham and Gubbins, Mo and Gubbins, Grunberg Nissan, and Di-Pippo[12, 15-17]. Most of the viscosity predictions for liquid mixture based on empirical and semi-theoretical methods are extensively employed. Empirical viscosity methods use mixing rules and/or parameters correlated with mixture ratio-viscosity or pressure-viscosity or temperature-viscosity equations[10-11,18-24]. Semi-theoretical models have a sound theoretical background, but the parameters are obtained from experimental data. The model requires minimum and easily obtainable data. Semi-theoretical models use one of the below theories; the principle of corresponding states, statistical mechanics model, reaction rate theory, square well theory, Lennard-Jones, hard sphere theory or their modifications[15-16]. Ely and Hanley[25] suggested the extended corresponding states approach. According to the principle of corresponding states, a dimensionless property of one substance is equal to that of another(reference) substance when both are evaluated at the same reduced conditions. Assael et al[4-5] applied Dymond's approach to the Enskog hard sphere theory. The viscosity measurements of binary n-heptane+n-alkane mixtures were performed with the vibrating-wire instrument by Assael et al. The accuracy of viscosity data was estimated to be ±0.5%. In this work, predicting viscosities of binary n-alkane mixtures would be compared with excellent experimental measurements mentioned above.

The theoretical models for gas or vapor phase viscosities are based on the kinetic theory of gases[12, 27-28]. There are very few theoretical approaches for the prediction of the viscosity of liquid mixtures. However, in several recent papers[17, 29-33], the theoretical approaches have been used for n-alkane mixtures. One of the theoretical methods was based on hard sphere theory of Dymond-Assael and Vesovic-Wakeham scheme[17, 29-31]. Other method used was Erying kinematic viscosity model for binary mixtures utilizing two-parameter van der Waals cubic equation of state[32]. Another method used was friction theory(f-theory) for viscosity modeling. f-theory was used cubic equation of states (SRK, PR, and PRSV) and Amontons-Coulomb friction law[33].

Our prediction methods for viscosity were based on empirical and semi-theoretical methods. The proposed empirical method employs a modified version of the Andrade equation[18-24, 34-35] to predict viscosities of binary n-alkane mixtures. The semi-theoretical method has a sound theoretical background. This method requires minimum and easily obtainable data as input. The proposed new semi-theoretical method based on both the Enskog's hard sphere theory for dense fluids and the principle of corresponding

states was successfully applied for predicting viscosity and thermal conductivity of pure organic liquids. In our previous studies, we obtained viscosities of pure organic liquids(C_5-C_{20}) and light petroleum fractions(C_5-C_{15}). The average absolute deviations were 2% for 75 pure hydrocarbon and 42 petroleum fractions. Details are presented elsewhere[36-38].

2. EMPIRICAL AND SEMI-THEORETICAL METHODS

In this study, empirical and semi-theoretical methods are presented for predicting viscosity of binary mixtures of n-heptane with n-hexane and n-nonane at eight different compositions.

2.1 Empirical Viscosity Method

In this work, the viscosity of mixture was calculated via Eq.(1)

$$\eta_{mix}=\sum w_i \eta_i \quad (1)$$

The critical properties (T_c,P_c), the molecular weight(M), and normal boiling point(T_b) were calculated according to simple combination rule.

$$T_c =\sum w_i T_{ci} \quad (2)$$

$$P_c =\sum w_i P_{ci} \quad (3)$$

$$M =\sum w_i M_i \quad (4)$$

$$T_b =\sum w_i T_{bi} \quad (5)$$

w is weight fraction for component i. Here, the empirical viscosity method, developed for binary n-alkane mixtures based on the Andrade[18-19] equation utilizing T_r and η^* instead of T and η respectively, is used.

$$\log\eta^* = -A + B/T_r \quad (6)$$

η^* is reduced dynamic viscosity, converted to dynamic viscosity via Eq.(7).

$$\eta=\eta^* T_c^{-1/6} M^{1/2} P_c^{-2/3} \quad (7)$$

where T_r is reduced temperature, T/T_c. The parameters A and B were calculated for 75 organic liquids separately using as many experimental viscosity-temperature data points as available. Details are presented elsewhere[37].

2.2 Semi-Theoretical Viscosity Method

A new semi-theoretical method based on Enskog's hard sphere theory for dense fluids and the principle of corresponding states was proposed and successfully applied for predicting the viscosity of pure organic liquids and petroleum fractions. Details of the methodology are presented elsewhere[37-38].

This work is based on the following model for dynamic viscosity of liquids: at a given reduced temperature and pressure, reduced dynamic viscosity, η^* can be calculated by adding a correction term to the reduced dynamic viscosity of a simple monatomic liquid at the same reduced conditions. At low and moderate pressures, the effect of pressure on liquid dynamic viscosity can be neglected. According to the proposed method[37-38] reduced dynamic viscosity of liquid mixtures(η^*) at a given reduced temperature(T_r) and 0.1 MPa, can be calculated by adding a correction term(η^*_f) to the reduced dynamic

viscosity term of pure organic liquids(η^*_m). η^* is determined as follows:

$$\eta^*(T_r . \theta) = \eta_m^* (T_r) + \eta^*_f (T_r . \theta) \quad (8)$$

where η^*_m is the reduced dynamic viscosity of monatomic liquid and η^*_f is a correction term which employs θ parameter. θ was selected as the best choice for all organic liquid groups where θ is the reduced temperature at the normal boiling point. θ is calculated for mixtures as follows:

$$\theta = \sum w_i T_{bi} / \sum w_i T_{ci} \quad (9)$$

In our work, η^*_m was developed for n-alkanes.

$$\eta_m^* = 10^{-9}[6.799 - 8.760 T_r + 2.816 T_r^2] \quad (10)$$

Although it is developed for n-alkanes, Eq.(10) proved to be satisfactory for other kinds of organic liquids and their mixtures as well. $\eta^*_f (T_r . \theta)$ function was determined for alkanes according to the following equations.

$$\eta^*_f (T_r . \theta) = \eta'_f (T_r) . \eta''_f (\theta) \quad (11)$$

$$\eta^*_f (T_r . \theta) = 0.0271\, T_r^{-5.285} [-130.5 + 720.5\theta - 1329\theta^2 + 825.1\theta^3] \quad (12)$$

The correlation coefficients are 0.9953, 0.9921, respectively. The reduced dynamic viscosity(η^*) of pure organic liquids is calculated using Eqs. (8-12). Reduced dynamic viscosity converted to dynamic viscosity via Eq.(7). Finally, the dynamic viscosity of binary n-alkane mixtures is determined according to Eq.(1). In this work, predicted viscosities of binary n-alkane mixtures were compared with excellent experimental data measured by Assael et al.[4]. The percentage of average absolute deviations were obtained by the following equation.

$$AAD\% = \left(\frac{|\eta_{calc} - \eta_{exp}|}{\eta_{exp}} \right) 100 \quad (13)$$

3. RESULTS

Table 1 presents the dynamic viscosities for binary mixtures of n-heptane with n-hexane and n-nonane at eight different compositions according to the above mentioned empirical new method. For 27 data points, average and maximum absolute deviations were 1.25% and 4.48%, respectively.

Dynamic viscosity of binary n-alkane mixtures were calculated according to the proposed semi-theoretical method. Results are presented in Table 2. Average and maximum absolute deviations were 0.87% and 2.63%, respectively. 27 experimental data points were utilized for eight different compositions of n-alkane mixtures.

Table 3 summarizes n-alkane liquid mixtures and calculated average absolute deviations with empirical and semi-theoretical methods. The average absolute deviations were 1.25% and 0.87% respectively, for 27 data points.

Empirical viscosity results are as accurate as or even better than the results of the other empirical methods covered in Table 4. Table 5 summarizes the average absolute deviation of selected pure organic liquid mixture viscosities according to several published theoretical and semi-theoretical methods. When compared with others published recently, the new empirical and semi-theoretical viscosity correlation yields better results than most of the other methods for mixtures.

4. CONCLUSION

In conclusion, for predicting viscosities of binary n-alkane liquid mixtures over a wide temperature range at atmospheric pressure, the correlation developed in this study should be preferred over all other such correlations. Utilization of both the principle of corresponding states and Enskog's hard sphere theory for dense fluids has lead to practical and accurate viscosity prediction formulae for mixtures.

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Table I. Empirical Viscosity Calculation Results at Atmospheric Pressure for the Eight Different Compositions of n-alkane Mixtures.

W_A	W_B	T(K)	$\eta(mPas)_{exp}$	M(kg/kmol)	P_c(Pa)	T_c(K)	T_b(K)	T_r	η^*_{calc}	η_{calc}(mPas)	AAD%
n-Heptane+n-Hexane											
0.4	0.6	303.15	0.3172	91.782	2875603.5	520.82	353.78	0.58206	1.0002E-04	0.31431	0.912
		323.15	0.2629	91.782	2875603.5	520.82	353.78	0.62046	8.3136E-05	0.26125	0.627
0.7	0.3	303.15	0.3440	95.991	2805689.3	530.51	362.69	0.57143	1.0883E-04	0.34299	0.294
		323.15	0.2842	95.991	2805689.3	530.51	362.69	0.60913	8.9578E-05	0.28232	0.661
n-Heptane+n-Nonane											
0.4	0.6	303.15	0.4999	117.03	2480436	572.60	403.04	0.52943	1.6370E-04	0.51810	3.642
		323.15	0.4024	117.03	2480436	572.60	403.04	0.56436	1.3008E-04	0.41172	2.316
0.7	0.3	303.15	0.4332	108.615	2608105.5	556.40	387.32	0.54484	1.3907E-04	0.44057	1.702
		323.15	0.3538	108.615	2608105.5	556.40	387.32	0.58079	1.1194E-04	0.35461	0.230
n-Heptane+n-Hexane											
0.4	0.6	293.353	0.3497	91.782	2875603.5	520.82	353.78	0.56325	1.1051E-04	0.34728	0.691
		303.332	0.3167	91.782	2875603.5	520.82	353.78	0.58241	9.9841E-05	0.31374	0.933
		312.261	0.2898	91.782	2875603.5	520.82	353.78	0.59956	9.1671E-05	0.28807	0.596
		323.144	0.2630	91.782	2875603.5	520.82	353.78	0.62045	8.3140E-05	0.26126	0.660
0.7	0.3	288.281	0.4034	95.991	2805689.3	530.51	362.69	0.54340	1.2799E-04	0.40339	0.002
		294.311	0.3776	95.991	2805689.3	530.51	362.69	0.55477	1.1961E-04	0.37696	0.169
		297.616	0.3644	95.991	2805689.3	530.51	362.69	0.56100	1.1538E-04	0.36364	0.208
		303.176	0.3439	95.991	2805689.3	530.51	362.69	0.57148	1.0880E-04	0.34290	0.292
		313.278	0.3114	95.991	2805689.3	530.51	362.69	0.59052	9.8305E-05	0.30983	0.504

		323.226	0.2838	95.991	2805689.3	530.51	362.69	0.60927	8.9516E-05	0.28213	0.589
n-Heptane+n-Nonane											
0.4	0.6	293.746	0.5585	117.03	2480436	572.60	403.04	0.51300	1.8436E-04	0.58351	4.478
		303.185	0.4997	117.03	2480436	572.60	403.04	0.52949	1.6363E-04	0.51788	3.639
		313.128	0.4479	117.03	2480436	572.60	403.04	0.54685	1.4543E-04	0.46028	2.765
		322.994	0.4032	117.03	2480436	572.60	403.04	0.56408	1.3030E-04	0.41241	2.285
0.7	0.3	293.965	0.4802	108.615	2608105.5	556.40	387.32	0.52833	1.5518E-04	0.49159	2.373
		303.411	0.4326	108.615	2608105.5	556.40	387.32	0.54531	1.3865E-04	0.43925	1.536
		312.673	0.3916	108.615	2608105.5	556.40	387.32	0.56196	1.2498E-04	0.39594	1.109
		323.128	0.3538	108.615	2608105.5	556.40	387.32	0.58075	1.1196E-04	0.35469	0.252
		333.254	0.3201	108.615	2608105.5	556.40	387.32	0.59895	1.0131E-04	0.32095	0.265
Total AAD%*											**1.249**

$$*\text{Total AAD\%} = \sum_{k=1}^{27} \left(\frac{\left| \eta_{calc} - \eta_{exp} \right|}{\eta_{exp}} .100 \right) \Big/ 27$$

Table II. Semi-Theoretical Viscosity Calculation Results at Atmospheric Pressure for the Eight Different Compositions of n-Alkane Mixtures.

W_A	W_B	T(K)	η_{exp}(mPas)	M (kg/kmol)	P_c(Pa)	T_c(K)	T_b(K)	T_r	$\theta(T_b/T_c)$	$\eta_m{}^*_{calc}$	η^*_{calc}	η_{calc}(mPas)	AAD%
n-Heptane+n-Hexane													
0.4	0.6	303.15	0.3172	91.782	2875604	520.82	353.78	0.5821	0.6793	2.655E-09	4.600E-09	0.3142	0.947
		323.15	0.2629	91.782	2875604	520.82	353.78	0.6205	0.6793	2.448E-09	3.836E-09	0.2620	0.334
0.7	0.3	303.15	0.3440	95.991	2805689	530.51	362.69	0.5714	0.6837	2.713E-09	4.991E-09	0.3419	0.612
		323.15	0.2842	95.991	2805689	530.51	362.69	0.6091	0.6837	2.508E-09	4.133E-09	0.2831	0.371
n-Heptane+n-Nonane													
0.4	0.6	303.15	0.4999	117.03	2480436	572.60	403.04	0.5294	0.7039	2.951E-09	7.426E-09	0.5108	2.183
		323.15	0.4024	117.03	2480436	572.60	403.04	0.5644	0.7039	2.753E-09	5.945E-09	0.4090	1.631
0.7	0.3	303.15	0.4332	108.615	2608106	556.40	387.32	0.5448	0.6961	2.863E-09	6.331E-09	0.4359	0.630
		323.15	0.3538	108.615	2608106	556.40	387.32	0.5808	0.6961	2.662E-09	5.136E-09	0.3537	0.040
n-Heptane+n-Hexane													
0.4	0.6	293.353	0.3497	91.782	2875604	520.82	353.78	0.5633	0.6793	2.759E-09	5.073E-09	0.3465	0.915
		303.332	0.3167	91.782	2875604	520.82	353.78	0.5824	0.6793	2.653E-09	4.592E-09	0.3136	0.965
		312.261	0.2898	91.782	2875604	520.82	353.78	0.5996	0.6793	2.560E-09	4.223E-09	0.2885	0.464
		323.144	0.2630	91.782	2875604	520.82	353.78	0.6205	0.6793	2.448E-09	3.837E-09	0.2620	0.367

0.7	0.3	288.281	0.4034	95.991	2805689	530.51	362.69	0.5434	0.6837	2.871E-09	5.842E-09	0.4002	0.795
		294.311	0.3776	95.991	2805689	530.51	362.69	0.5548	0.6837	2.806E-09	5.470E-09	0.3747	0.772
		297.616	0.3644	95.991	2805689	530.51	362.69	0.5610	0.6837	2.771E-09	5.282E-09	0.3618	0.704
		303.176	0.3439	95.991	2805689	530.51	362.69	0.5715	0.6837	2.713E-09	4.990E-09	0.3418	0.609
		313.278	0.3114	95.991	2805689	530.51	362.69	0.5905	0.6837	2.608E-09	4.523E-09	0.3098	0.501
		323.226	0.2838	95.991	2805689	530.51	362.69	0.6093	0.6837	2.507E-09	4.131E-09	0.2830	0.298
n-Heptane+n-Nonane													
0.4	0.6	293.746	0.5585	117.03	2480436	572.60	403.04	0.5130	0.7039	3.047E-09	8.332E-09	0.5732	2.627
		303.185	0.4997	117.03	2480436	572.60	403.04	0.5295	0.7039	2.951E-09	7.423E-09	0.5106	2.182
		313.128	0.4479	117.03	2480436	572.60	403.04	0.5469	0.7039	2.851E-09	6.622E-09	0.4555	1.703
		322.994	0.4032	117.03	2480436	572.60	403.04	0.5641	0.7039	2.754E-09	5.955E-09	0.4096	1.595
0.7	0.3	293.965	0.4802	108.615	2608106	556.40	387.32	0.5283	0.6961	2.957E-09	7.038E-09	0.4846	0.920
		303.411	0.4326	108.615	2608106	556.40	387.32	0.5453	0.6961	2.860E-09	6.313E-09	0.4347	0.476
		312.673	0.3916	108.615	2608106	556.40	387.32	0.5620	0.6961	2.766E-09	5.712E-09	0.3933	0.425
		323.128	0.3538	108.615	2608106	556.40	387.32	0.5807	0.6961	2.662E-09	5.137E-09	0.3537	0.019
		333.254	0.3201	108.615	2608106	556.40	387.32	0.5989	0.6961	2.563E-09	4.666E-09	0.3213	0.366
Total AAD%													**0.869**

Table III. Empirical and Semi-Theoretical Viscosity Calculation Results

n-Alkane Mixtures	Temperature Range(K)	Data Points	AAD% Empirical	AAD% Semi-Theoretical
n-Heptane+n-Hexane	288.28-323.23	14	0.510	0.618
n-Heptane+n-Nonane	293.75-333.25	13	2.046	1.138
Total	**288.28-333.25**	**27**	**1.249**	**0.869**

Table IV. Empirical Methods for Viscosity Predictions of Pure Organic Liquid Mixtures

Methods	Mixtures	Correlations	Data Points	AAD%
Lee(1966)	n-Metane-n-Decane	Simple Mixture Rule	217	<4
Katti(1966)	Benzene-Cyclohexane, Carbontetrachloride- Cyclohexane, Carbontetrachloride- Benzene	Grunberg-Nissan	26	5.7-17.6
Mussche-Verhoeye(1975)	10 binary and 1 ternary	Mc-Allister	169	1-3
		Heric		4-5
Aasen(1990)	7 mixtures	Arrhenius - Doolittle	35	1.9-3.8
Serrano(1990)	3 binary and 1ternary	Mc-Allister	221	0.3-0.4
Alan and Teja(1991)	11 binary mixtures	Simple Mixture Rule, ECN	353	5.8
New Method(2002)	n-Heptane+n-Hexane, n-Heptane+n-Nonane	Simple Mixture Rule, Andrade	27	1.2

Table V. Theoretical and Semi-Theoretical Methods for Viscosity Predictions of Pure Organic Liquid Mixtures

Methods	Mixtures	Correlations	Data Points	AAD%
Ely-Hanley(1981)	26 binary	ECSP	455	7
Teja-Rice(1981)	29 binary	ECSP	1010	3.8
Wei-Rowley(1984)	24 binary	NRTL	191	1.6-5.2
Ducoulombier(1986)	4 binary and 1 ternary	ECSP	63	10.6
Assael(1992a)	3 binary	EHST	114	<5
Assael(1992b)	32 binary, 2 ternary, 3 quaternary	EHST	1284	<5
Assael(2000)	n-Hexane-Toluene	Dymond-Assael	21	7
	n-Hexane+Cyclohexane	Vesovic-Wakeham	14	5
Assael(2001)	n-Hexane+n-Heptane	Dymond-Assael	35	9.5
		Vesovic-Wakeham		
Lee(2001)	43 n-Alkane mixtures ,	Van der Waals EOS	104	1-3
	61 Aromatics and n-Alkane mixtures			
Cisneros(2001)	12 binary, 1 ternary, 1 quaternary	f-theory	-	4
New Method(2002)	n-Heptane+n-Hexane,	EHST and ECSP	27	0.9
	n-Heptane+n-Nonane			

ECSP: Extended Corresponding States Principle

EHST: Enskog's Hard Sphere Theory

EOS: Equations of States

NRTL: Nonrandom two-liquid